Evaluating the Impact of Municipal Water Fluoridation on the Aquatic Environment

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Abstract: Although highly beneficial for dental health, low concentrations of fluoride in environmental waters may be toxic to several organisms. In an era of heightened public awareness about the environment, this may lead city officials to withhold implementing water fluoridation for environmental reasons. This paper presents a mass balance approach to evaluate this perceived risk. Generally speaking, fluoridated water loss during use, dilution of sewage by rain and ground water infiltrate, fluoride removal during secondary sewage treatment, and diffusion dynamics at effluent outfall combine to eliminate fluoridation-related environmental effects. In Montreal, water fluoridation would raise average aquatic fluoride levels in the waste water plume immediately below effluent outfall by only

0.05–0.09 mg/l. Downstream, these changes would be only 0.02–0.05 mg/l at 1 km, and 0.01–0.03 mg/l at 2 km below outfall. Overall river fluoride concentrations theoretically would be raised by 0.001–0.002 mg/l, a value not measurable by current analytical techniques. All resulting concentrations would be well below those recommended for environmental safety and would not exceed natural levels found elsewhere in Quebec. A literature review did not reveal any examples of municipal water fluoridation causing recommended environmental concentrations to be exceeded, although excesses occurred in several cases of severe industrial water pollution. (Am J Public Health 1990; 80:1230–1235.)

Introduction

The beneficial effects of water fluoridation for human health are widely recognized. 1-3 Optimally fluoridated drinking water significantly reduces dental caries in both children and adults. 2-5 Fluoride may also reduce the incidence of osteoporosis and hip fractures, although the evidence is controversial. 6-8 No side effects occur. 1-3 Although the prevalence of mild dental fluorosis may increase, the esthetic effects of this mild tooth whitening are greatly outweighed by the improved esthetics of fewer decayed, missing, and filled teeth. 9 By far the most cost-effective intervention in dentistry, 10 optimally fluoridated water is provided in more than 10,000 North American communities and in countries around the globe. 11-13

The toxic environmental effects of fluorides are also well documented, however. Airborne fluoride pollution from industrial sources may cause severe effects in plants by inhibiting photosynthesis, chlorophyll production and carbohydrate metabolism, with resulting defoliation or death.^{14–16} Most plants accumulate fluoride which may lead to bony and dental fluorosis, lameness, arthritis, and other symptoms in wild and domestic herbivores foraging downwind from fluoride air pollution sources.¹⁷

As a water pollutant, elevated concentrations of fluoride may affect a number of organisms, including fish, amphibians, insects, snails, shellfish, protozoa, and some aquatic plants. Fresh water fish, especially rainbow trout and other trout species, appear to be particularly sensitive and may show growth and behavioral changes, decreased survival, and prolonged hatching time when exposed to moderate levels. Under negligible water hardness conditions, lethal effects for rainbow trout may occur at fluoride concentrations as low as 2.7–4.7 mg/l²⁰ and one undocumented observation

reported delayed hatching of trout eggs at 1.5 mg/l.²¹ Small amounts of calcium carbonate found in soft or moderately hard water, however, greatly reduce these effects,²² as do water hardness generally, alkalinity, the presence of metallic ions (particularly calcium and magnesium) and chloride.^{22–24} Toxicity increases with water temperature, by increasing fish metabolism.²⁵

To protect aquatic organisms, several authorities recommend an upper limit between 1.0 mg/l and 1.8 mg/l of fluoride for fresh water systems. 18,26-28 Others feel that the environmental literature on fluoride toxicity is incomplete. They recommend that levels protecting drinking water (generally 1.2 to 2.0 mg/l) also be used to protect the environment, arguing that detectable effects occur in humans at lower concentrations than in other species. 29-32 Nevertheless, some states allow concentrations as high as 5-10 mg/l if the contaminated water is not subsequently used for human consumption. 33 No effluent standards exist.

Industrial waste and municipal sewage may add to the fluoride which is naturally present in all surface water.³³ In an era of heightened public awareness about environmental issues, concerns of public interest groups may lead city officials and politicians to withhold water fluoridation for environmental reasons, as occurred in Montreal in 1988.

To evaluate the environmental impact of water fluoridation, this paper uses a mass balance (material balance) approach to develop a series of mathematical equations which describe the fate of fluoride added to drinking water in a typical municipal water management system. In this approach, the ionic mass of fluoride entering the aquatic system from all sources is calculated, its distribution followed, and its fate examined; by balancing fluoride inputs and outputs, intervening concentrations and changes may be determined. This approach, widely used in environmental impact analyses,³⁴ is analagous to the study of physiologic or pharmacologic agents in living organisms.

The equations described below may be applied to readily available data from relevent water authorities to predict potential fluoridation-related changes in environmental waters receiving municipal effluent. These changes may be compared to natural variations in local ambient fluoride concentrations, the scientific literature concerning fluoride toxicity to aquatic organisms, and to recommendations from

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scientific and government agencies. The city of Montreal is presented as an example.

Methods

Fate of Fluorides in Municipal Water Systems

For most municipal water management systems, drinking water is drawn from a lake, river, or subterrain well and is pumped to a purification plant. There it may undergo aeration, flocculation, coagulation and sedimentation, filtration and be disinfected with ozone or chlorine dioxide. Usually a small amount of residual chlorine is maintained in the drinking water to avoid bacterial regrowth in the distribution system. Ambient fluoride concentrations are generally measured in the final stages of water purification. Fluoride is then added to obtain optimal levels, between 0.7 and 1.2 mg/l depending on climatic conditions. Although several fluoridecontaining compounds may be used, all dissociate on admixing, thereby liberating free fluoride ion (F-).35 Occasionally, when drinking water sources contain high concentrations of fluoride, the water may be "defluoridated" to optimal levels by one of several methods.36

Drinking water is distributed to a variety of residential. industrial, commercial, and municipal clients. Waste water from all these sources is collected by the city sewer system and is transported to the sewage treatment plant. Industrial and human wastes increase sewage fluoride levels. Some fluoride is lost, however, when drinking water is used to water gardens, wash cars, fill pools, extinguish fires, etc. Infiltrating ground water, underground streams, and rain may add to sewage flow and cause considerable dilution.

Primary sewage treatment removes suspended solids and phosphates by screening, filtration, flocculation, coagulation and sedimentation. Several common processes using iron salt coagulation, lime, or activated alumina may remove fluoride at high concentrations; however, these effects are minor at the low concentrations of fluoride found in municipal sewage.36,37 After initial treatment, waste water undergoes secondary (biological) treatment in order to destroy unwanted organisms and reduce nitrogen levels and biochemical oxygen demand (BOD). The microorganisms present in most secondary treatment systems absorb large quantities of fluoride and may reduce fluoride concentrations in effluent by up to 50 percent.38,39 Effluent may then undergo further disinfection with chlorine before discharge.

Effluent is generally discharged from an outfall pipe at a lake, river, or ocean bottom. The warmer effluent rises quickly to the surface creating physical turbulence and an immediate dilution which may be as high as 5-15 times. If natural dilution is low, a diffusion apparatus may be installed at effluent outfall which distributes effluent evenly over a wide area to ensure similar dilution levels. 40 Natural currents then carry the waste water downstream, creating a waste water "plume" and secondary dilution over several miles.

Mathematical Equations Evaluating Fluoridation Impact

From this overview of the fate of fluorides used in water fluoridation, mathematical equations may be derived to estimate the impact of fluoridation on the aquatic environment downstream of municipal sewage outfall. Terms used in these equations are defined in Table 1.

Global Impact

An estimate of global environmental impact may be obtained by comparing the fluoride needed to optimally fluoridate drinking water to the amount naturally present in

TABLE 1—Definition of Symbols

Fluoride Concentrations'

fluoride concentration in municipal drinking water

F_M F_R F_T fluoride concentration in raw sewage fluoride concentration in treated sewage

fluoride concentration in environmental water receiving municipal effluent

 $\mathbf{F}_{\mathbf{X}}$ fluoride concentration in waste water plume at point X,

downstream from effluent outfall site

Flow Rates

flow rate (production) of municipal drinking water

flow rate of sewage at treatment plant

flow rate of environmental water receiving municipal effluent

portion of municipal drinking water destined for sewers

portion of sewage originating as drinking water

Q_M = Q_S = Q_E = Portions P_M = P_D = PE_X = portion of environmental water affected by waste water plume at point X

Diffusion Factor

a unitless number describing cumulative dilution occurring in the waste water plume from effluent outfall to point X downstream

*Fluoride concentrations before and after fluoridation (see text) are respectively indicated by subscripts 1 and 2.

the aquatic system receiving municipal effluent. Adjusted fluoride equals the difference between optimal and preexisting drinking water fluoride concentrations (ΔF_M) multiplied by the rate per unit time of drinking water production (Q_M). Environmental fluoride equals the sum of fluoride naturally present in drinking water (F_{M1}Q_M) and fluoride naturally present in the waters receiving municipal effluent (F_{E1}Q_E). Thus,

$$\frac{\text{Municipal F}}{\text{Environmental F}} = \frac{\Delta F_{M} Q_{M}}{F_{El} Q_{E} + F_{Ml} Q_{M}}$$
(1)

If Q_M is very small relative to Q_E , in the case of a small municipality discharging into a large river, then Equation 1 simplifies to:

$$\frac{\text{Municipal F}}{\text{Environmental F}} = \frac{\Delta F_{\text{M}} Q_{\text{M}}}{F_{\text{EI}} Q_{\text{E}}} \tag{2}$$

Multiple municipalities and river systems may be considered in an additive fashion using the general formula:

$$\frac{\text{Municipal F}}{\text{Environmental F}} = \frac{\Sigma_{i} \Delta F_{Mi} Q_{Mi}}{\Sigma_{i} F_{E1i} Q_{Ei} + \Sigma_{i} F_{M1i} Q_{Mi}}$$
(3)

The change in environmental fluoride concentrations due to fluoridation (ΔF_E) may be obtained by dividing the fluoride added to drinking water by the sum of municipal and receiving water flow rates.

$$\Delta F_{E} = \frac{\Delta F_{M} Q_{M}}{Q_{E} + Q_{M}} \tag{4}$$

In a multiple city or river situation, this relationship becomes:

$$\Delta F_{E} = \frac{\Sigma_{i} \Delta F_{Mi} Q_{Mi}}{\Sigma_{i} Q_{Ei} + \Sigma_{i} Q_{Mi}}$$
 (5)

Obviously, the post-fluoridation concentration in water downstream from municipal sewage outfall equals the sum of the calculated change and the pre-existing concentration. The ratio of these two values $(\Delta F_{\rm E}/F_{\rm E1})$ is identical to Equation 1

While conceptually simple, these models overestimate the real effects of fluoridation on the environment by assuming that all drinking water is discharged as waste, by excluding the effects of sewage treatment, and by discounting eventual fluoride deposition into sediments. Moreover, estimates from these models are valid only at some theoretical point downstream where municipal effluent and receiving waters have become thoroughly mixed.

The first of these objections may be overcome by including in the numerator of Equations 1-5 only the percentage of drinking water which is eventually discharged as effluent (P_M) . Thus, for example, Equation 1 would be corrected to:

$$\frac{\text{Municipal F}}{\text{Environmental F}} = \frac{\Delta F_{M} Q_{M} P_{M}}{F_{EI} Q_{E} + F_{MI} Q_{M}}$$
 (6)

Local Impact

To estimate fluoridation-related changes in fluoride concentrations in the waste water plume downstream from municipal effluent outfall, the effect of sewage treatment must first be evaluated by comparing fluoride concentrations in treated (F_{T1}) and raw (F_{R1}) sewage. The ratio of these values (F_{T1}/F_{R1}) shows the portion of fluoride remaining after sewage treatment and is not affected by fluoridation status. ^{38,39} The concentration of fluoride in post-fluoridation treated municipal effluent (F_{T2}) may then be estimated.

$$F_{T2} = F_{T1} + \frac{\Delta F_M P_D F_{T1}}{F_{R1}}$$
 (7)

 $P_{\rm D}$ is the portion of sewage originating as drinking water and equals $Q_{\rm M}P_{\rm M}/Q_{\rm S}$. From the rate of diffusion of waste water in the effluent plume $(DF_{\rm X})$, the concentration of fluoride in the plume $(F_{\rm X2})$ may now be estimated at outfall and for points downstream.

$$F_{X2} = \frac{F_{T2} + F_{E1}(DF_X - 1)}{DF_Y}$$
 (8)

The change in fluoride concentration at any point downstream from outfall (ΔF_x) equals:

$$\Delta F_{X} = \frac{\Delta F_{T}}{DF_{X}} \tag{9}$$

Waste water is unequally distributed in the plume. Thus, diffusion, and DF_{X} , is greatest at the edge where effluent concentrations are least and, conversely, least at the center where concentrations are highest. Also, turbulence may create eddies of poorly diluted effluent at the outfall site, which disappear downstream. It should be noted that DF_{X} has a maximum value equal to flow in the receiving water (Q_{E}) divided by flow of municipal sewage (Q_{S}) ; generally this limit is important only when large volumes of effluent are discharged into small bodies of water.

The effluent plume affects only a portion of the receiving waters. This portion (P_X) at point "X" may be calculated as follows:

$$P_{X} = \frac{Q_{S}(DF_{X})}{O_{F}} \tag{10}$$

Results

An Example: The City of Montreal

Montreal's drinking water, serving 1,6 million people, is drawn from the St. Lawrence River, near the Lachine Rapids slightly upstream from the city, and is returned some 35 km downstream at Ile-aux-Vaches. From data provided by municipal, provincial, and federal agencies, 41-47 potential global changes in fluoride levels related to water fluoridation were estimated (Table 2). At the Lachine Rapids the daily water flow of the St. Lawrence averages 883 (range 659–1028) million cubic meters, and natural fluoride levels average 0.13 (range 0.11-0.17) mg/l. On average, therefore, 115 tonnes (tonne = 1000 kg = 2,204.6 pounds) of naturally present fluoride (theoretical range 73-175) flow past Montreal daily. Assuming all municipal drinking water eventually returns to the river, fluoridation would add an additional 1.93 tonnes to this water, or 1/60 (1.7 percent) of naturally occurring amounts. On average, however, only 57 percent of Montreal's drinking water returns to the river, making the additional fluoride load closer to 1.09 tonnes, or 1 percent of natural amounts. This additional charge is well within natural variations in fluoride concentration (±15 percent), river flow (\pm 16 percent), and fluoride ion mass (\pm 37 percent). Overall, the average concentration of fluoride in the river downstream from effluent outfall would increase by 0.001-0.002 mg/l, a value which is not measurable by current analytical methods.

Excluding rain, Montreal produces an average of 2.4 million cubic meters of sewage per day, of which 42 percent originates as unfluoridated drinking water, 2 percent as fluoridated drinking water from neighboring suburbs, and 56 percent from infiltrating ground water. This latter value is very high, due to Montreal's island geography, and an old combined sewer system in one sector. Optimal fluoridation in Montreal would increase drinking water fluoride concentrations from 0.11-0.15 mg/l to 1.2 mg/l which would in turn increase the average concentration of fluoride in Montreal sewage by 0.45 ± 0.01 mg/l. Daily fluoride concentrations in

TABLE 2—Average Daily Water Flow and Fluoride Levels in the St. Lawrence River and Montreal Water Supplies, before and after Fluoridation (range in parentheses)

	Flow Rate (m ³ × 10 ⁶)	Fluoride Ion Concentration (mg/l)	Fluoride Ion Mass (metric tons)
St. Lawrence	883	0.13	115
River	(659–1028)	(0.11–0.17)	(73–175)
Water supply before fluoridation	1.8	0.13	0.23
	(1.5–2.2)	(0.11–0.15)	(0.16–0.33)
Water supply after fluoridation	1.8 (1.5–2.2)	1.2	2.16 (1.80–2.64)
Fluoride added to	1.8	1.07	1.93
water supply	(1.5–2.2)	(1.05–1.09)	(1.64–2.31)
Fluoride added to waste water from water supply	1.02*	1.07 (1.05–1.09)	1.09 (1.02–1.11)

^{*}Range not available

Montreal sewage average 0.34 ± 0.07 mg/l (mean \pm SD) and range from 0.22 to 0.58 mg/l.⁴⁵ Thus, after fluoridation, concentrations in Montreal sewage would become 0.79 \pm 0.08 mg/l (mean \pm SD) and range from 0.66 to 1.04 mg/l. Because Montreal sewage does not undergo biological treatment, little or no fluoride removal occurs at the sewage treatment plant.

The diffusion of Montreal's waste water in the St. Lawrence river was measured in 1983 as part of a feasibility study for the new municipal sewage treatment plant.46 At sewage outfall a minimum initial diffusion factor of 5 occurred at the center of the waste water plume, and initial diffusion averaged 15 over the plume's entire cross-sectional area. After adjusting for current modifications which will increase outfall capacity and thereby decrease diffusion slightly, an average increase of 0.05 mg/l in fluoride concentration would be predicted across the plume immediately below effluent outfall (Table 3) and an average increase of 0.09 mg/l at the plume's center (Table 4). The plume's average concentration immediately below outfall would be 0.20 mg/l and the average level in the plume center at outfall would be 0.26 mg/l. Given known variations of fluoride ion concentrations in Montreal's sewage, the greatest daily average concentration expected would 0.34 mg/l at the outfall center. These values would decrease rapidly downstream. At 1 km from outfall an increase of only 0.02-0.05 mg/l would be expected. The predicted change at 2 km would be only 0.01-0.03 mg/l and would affect only 3.5-6.3 percent of the river.

Discussion

Using a mass balance approach, mathematical equations may be derived to evaluate the environmental impact of fluoride added to optimally fluoridate municipal water supplies. Generally, it will be shown that fluoridation has little impact on the surrounding aquatic environment. Some fluoride is lost when drinking water is used to water lawns, fill pools, etc; this usually ends up in the soil which may already contain large amounts (100-300 ppm).3,33 Although fluoride concentrations in the sewer system may increase with the addition of industrial and human wastes, they are diluted by rain and infiltrated ground water. Up to 50 percent of fluoride may be removed during secondary sewage treatment. Initial diffusion rapidly dilutes the remainder at the effluent outfall site. Secondary diffusion causes further reductions downstream. Eventually, the remaining fluoride is incorporated into river sediments, or carried to the ocean which already contain large amounts (1.2–1.4 mg/l).³³ Any toxicity from this residual fluoride would be severely inhibited by other sub-

TABLE 3—Estimated Average Daily Fluoride Concentrations (mg/l) across Effluent Plume before and after Municipal Water Fluoridation in Montreal (range in parentheses)

Distance from Outfall (km)	Diffusion Factor*	Concentration before Fluoridation	Concentration after Fluoridation	Change	Portion of River Affected %
0.3	10	0.15	0.20	0.05	2.0
1	19	(0.12–0.21) 0.14 (0.12–0.19)	(0.16–0.26) 0.16 (0.14–0.22)	0.02	3.9
2	31	0.14 (0.11–0.18)	0.15 (0.13–0.20)	0.01	6.3

^{*}Adjusted for current modifications to increase effluent capacity.

TABLE 4—Estimated Average Daily Fluoride Concentrations (mg/l) at Effluent Plume Center before and after Municipal Water Fluoridation in Montreal (range in parentheses)

Distance from Outfall (km)	Diffusion Factor*	Concentration before Fluoridation	Concentration after Fluoridation	Change	Portion of River Affected %
0.3	5	0.17 (0.13–0.25)	0.26 (0.22–0.34)	0.09	1.0
1	10	0.15 (0.12–0.21)	0.20 (0.16–0.26)	0.05	2.0
2	17	0.14 (0.12–0.19)	0.17 (0.14–0.22)	0.03	3.5

^{*}Adjusted for current modifications to increase effluent capacity.

stances present in surface and waste waters, notably water hardness, calcium ion, and chloride.²²⁻²⁴

Montreal is a typical example. Due to its island geography, infiltrating ground water in the sewer system is probably greater than elsewhere, but this effect is offset by the lack of a secondary sewage treatment plant. Water fluoridation in Montreal would result in a slight increase in the fluoride concentration of the waste water plume directly below the effluent outfall. These changes would return to pre-existing levels within two kilometers. Global changes would not be measurable. Rain water, not included in this analysis, would reduce sewage concentrations and these findings even further

The maximum predicted daily concentration at the center of the outfall site (0.34 mg/l) would be well below recommended levels for environmental safety, 18,26-32 and would be similar to natural levels occurring elsewhere. A survey in Quebec showed that average daily fresh water fluoride levels across the province varied from 0.07 to 0.24 mg/l and maximum levels of 1.7 mg/l occurred.⁴⁷ The average concentration in US rivers measured at 343 stations of the National Stream Quality Accounting Network in 1975 was 0.33 mg/l⁴⁸; six streams in the southwest US had concentrations of 1.4 to 1.8 mg/l. Other investigators report that fluoride concentrations in US rivers average 0.2 mg/l and range from 0.0 to 6.5 mg/l.49 In a polluted section of the Illinois River around Peoria, fluoride concentrations averaged 1.08 mg/l and ranged from 0.17 to 2.06 mg/l.50 The source of the pollution was unspecified. Ocean concentrations range from 1.2 to 1.4 mg/l.³³ One result is that marine and estuarine organisms are relatively immune to the toxic effect of fluoride.33,51

The predicted increase of 0.45 mg/l in the fluoride concentration of Montreal sewage due to water fluoridation is similar to findings elsewhere. Fluoride levels in raw sewage from seven fluoridated Ontario communities contained 0.47 mg/l more fluoride than raw sewage from 11 non-fluoridated communities (0.96 vs 0.49 mg/l), and treated sewage differed by 0.33 mg/l (0.62 vs 0.29 mg/l).³⁸ For 56 California cities, raw and primary treated sewage from fluoridated communities contained an average of 1.80 mg/l (range 1.4-3.2) or 0.37 mg/l more fluoride than non-fluoridated communities (average 1.43; range 0.6-3.6 mg/l).³⁹ Both studies showed the important effects of secondary sewage treatment which reduced effluent fluoride concentrations by 35 percent in Ontario and by 56 percent in California in both fluoridated and nonfluoridated communities. In California, domestic waste accounted for far more sewage fluoride than water fluoridation.

The fate of fluoride in waste water from municipal and industrial sources has been evaluated by a number of researchers. In 1973, fluoridated Bozeman, Montana (population 20,000) discharged an average of 8.5 m³/min of primary treated municipal effluent into the East Gallatin River (average flow: 146.7m³/min). This raised average fluoride levels from 0.33 mg/l, nine meters above outfall, to 0.62 mg/l at 0.3 km downstream.⁵² A maximum level of 2.0 mg/l was recorded. Natural fluoride levels were reestablished 5.3 km downstream. Another study at the same site found that fluoride concentrations quadrupled at effluent outfall but returned to natural levels 4 km downstream.⁵³ These changes were not considered to have any effect on the environment.²⁹

In 1977, fluoridated Minneapolis-St. Paul produced effluent at the rate of 9.6 m³/sec with a fluoride concentration of 1.21 mg/l, which was added to the Mississippi River (flow rate 279 m³/sec). Water collected 10 air miles below outfall contained the same fluoride concentration as that collected upstream from the sewage disposal plant.⁵⁴

In 1986, Kudo, et al, 55 evaluated fluoride concentrations along four small rivers in an industrialized area of the French Alps near unfluoridated Grenoble. This city (population 350,000) added about 1.8 tonnes of fluoride daily in untreated sewage to the Isere River (flow rate 300 m³/sec) which increased fluoride concentrations from 0.19 mg/l above the city to 0.26 mg/l some 70 km downstream. This study underlined the importance of industrial water pollution sources. On the small Arc River (flow rate 50 m³/sec) fluoride concentrations increased from 0.13 mg/l to 0.62 mg/l over a 50 km portion during which it received industrial effluents from three aluminum factories, a factory producing silica, and a factory producing phosphates. Severe fluoride-containing air pollution from these companies had destroyed much of the area's vegetation.

In another study underlining the importance of industrial water pollution, Martin and Salvadori⁵⁶ found that industrial effluents from a fluorophosphate fertilizer plant in Rouen, France increased fluoride concentrations in the Seine River from 0.25 to 2.0 mg/l which eventually fell to 0.47 mg/l many miles downstream. Fluoride discharge was estimated to be 37 tonnes per day, a quantity which would fluoridate a city 20 times the size of Montreal!

In conclusion, by using a mass balance approach, fluoridation-related changes in environmental concentrations of fluoride may be estimated from knowledge of municipal water management systems and data which are usually readily available from appropriate water authorities. Generally speaking, these changes will be minimal and, except when accompanied by serious industrial pollution, will remain below toxic levels recorded in the literature and recommendations by scientific authorities for the protection of the environment and human health.

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